# **Observed trends in stratospheric NO<sub>2</sub>**

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The goal of this study is to analyze linear trends in stratospheric column NO<sub>2</sub> contents as function of latitude using updated data of ground-based spectrometric (visible) measurements. Similar results derived from shorter data series were first reported by Gruzdev (2007, 2008). Trend analysis of column NO<sub>2</sub> at individual sites was earlier done by Liley et al. (2000), Gruzdev and Elokhov (2002), Gruzdev et al. (2004), Synyakov et al (2004).

#### DATA

Data used for the analysis are stratospheric column NO<sub>2</sub> contents measured at stations within the Network for the Detection of Atmospheric Composition Change – NDACC (previous NDSC – Network for the Detection of Stratospheric Change). Stations used are listed in Table. The data are publicly available on the web (ftp://ftp.cpc.ncep.noaa.gov/ndacc).

Observations are done in during morning and evening twilight and provide morning and evening series of  $NO_2$  contents corresponding to solar zenith angle 90°.

#### **ANALYSIS METHOD**

For trend analysis, a statistical multiple regression model used taking into account the annual variation, 11-year solar cycle, effects of El Chichon and Pinatubo eruptions, and effects of the quasi-biennial (QBO) end El Nino-Southern Oscillations (SO) (see Gruzdev, 2008, for more detail).

Figure 1 shows examples of approximation of the NO<sub>2</sub> time series by the regression model.

Spectral analysis shows that the residual series at some stations can contain weak periodic components of annual and intra-annual scales (Figure 2), which surely do not disappear after including additional members – harmonic functions of appropriate periods – in the statistical regression model. One probable reason is nonlinear interaction of quasiperiodic atmospheric processes with each other and with the annual cycle resulting in variations with combination frequencies, which cannot be accounted for in the framework of a linear model.

### RESULTS

#### **Annual linear trend estimates**

Figure 3 shows the latitudinal dependence of the annual estimates of the NO<sub>2</sub> linear trends for complete periods of observations. This figure reveals features peculiar for different latitude bands and regions in the southern (SH) and northern (NH) hemispheres. The annual trends statistically significant at 95% level are observed in the middle latitudes of the two hemispheres. These trends are positive in the SH middle latitudes but negative in the European sector of the NH middle latitudes, with module values generally within 6-12% per decade. In the Middle Asia (~ 40°N), the positive annual trend of smaller magnitude is observed. The annual trend estimates in the low, high and polar latitudes of the two hemispheres are usually statistically insignificant except in the NH high latitudes (67-68°N) where positive trends ~7% per decade are noted in Eastern Siberia and negative trends of about -2-4% per decade can be observed in Europe.

Figure 3 shows also the trend in stratospheric column NO<sub>2</sub> calculated with the help of 2-dimensional model SOCRATES (see e.g. Gruzdev and Brasseur, 2007; Gruzdev, 2008). The model calculations took into account observed trends in surface concentrations of  $CO_2$ ,  $CH_4$ ,  $N_2O$ , and CFCs, and in surface temperature. Figure 3 shows that, at least at two-thirds of the stations, there is a significant disagreement between the observed and calculated trends.

Effects of the increase in  $N_2O$ , the decrease in stratospheric ozone and temperature on stratospheric  $NO_2$  trends were studied analytically

(Gruzdev, 2008). The analysis has shown that the total effect is sensitive to concrete conditions but is expected to be of positive or negative trend of small magnitude (~1% per decade or less). This finding is in a good agreement with the results of 2-D model calculations. The discrepancy between the observed and calculated NO<sub>2</sub> trends may in particular be due to regional (longitude) dependence of the observed trends which is not taken into account in the used models.

## Seasonal trends

Figure 4 shows the seasonal estimates of the NO<sub>2</sub> trends. Seasonal trend estimates can differ from the annual estimates. For example, in winter and spring, statistically significant NO<sub>2</sub> trends are observed in the SH and NH low latitudes (~20°), positive in the SH and negative in the NH. Large positive trend of about 17% per decade is observed in spring in the Antarctic (~78°S). Summer trends are usually positive in the high latitudes of the two hemispheres (60-68°). Unlike at other stations, the signs of the seasonal trends at midlatitude stations of Lauder (New Zealand), Jungfraujoch and Zvenigorod (the both in Europe) do not depend on season and coincides with the signs of the annual trends (positive in the SH and negative in the NH).

## Solar cycle effect

Figure 5 shows a latitudinal distribution of the NO<sub>2</sub> change from the minimum to maximum phase of the 11-year cycle of solar activity (SA) according to morning measurements. In the middle latitudes of the two hemispheres, the stratospheric NO<sub>2</sub> content is, on the whole, larger during the SA minimum smaller than during the SA maximum. The difference can approach 9%. In the middle latitudes of the both hemispheres, the SA effect decreases (by module) with decrease in latitude. In the NH, the SA effect changes its sign to positive in the vicinity of 40°N.

The negative SA effect in  $NO_2$  is also noted at the Antarctic station of Dumont d'Urville that is located in the same longitudinal sector with the nearest midlatitude station on the Macquarie Island.

A significant negative SA effect in  $NO_2$  is noted at the stations that are located in the regions with a significant positive SA effect in total ozone (Gruzdev and Brasseur, 2007).

Figure 5 shows also the SA effect in stratospheric column  $NO_2$  derived from calculations with the help of two-dimensional model SOCRATES (Gruzdev and Brasseur, 2007). On the whole, the calculated effect corresponds in sign to observations but is several times smaller than the effect observed at some stations. One probable reason of this disagreement is a regional character of the SA effect which cannot be accounted for in a 2D model.

## Effect of volcanic aerosol

The maximum (in absolute value) decrease in stratospheric column  $NO_2$  after the Mt. Pinatubo eruption is shown in Fig. 6 according to morning and evening measurements. These maximum decrease values were observed in summer period in the NH in 1992 and in the SH in 1991/1992.

It is interesting that the relative (percent) decrease of evening  $NO_2$  contents corresponding to the maximum absolute decrease is approximately independent on latitude (within 20-25%), while the decrease in morning contents varies with latitude with maximum values of 32% at Lauder in the SH and at Zvenigorod in the NH.

## **CONCLUDING REMARKS**

Estimates of the linear trends and the solar activity effect in stratospheric  $NO_2$  will apparently be refined with increasing period of measurements.

Changes in stratospheric  $NO_2$  are related in particular to changes in stratospheric concentrations of  $N_2O$  and ozone and in stratospheric temperature. The mechanisms of influence of these factors on the  $NO_2$ content is such that the compensation of their effects in  $NO_2$  is possible, and this circumstance specifies the sensitivity of  $NO_2$  changes to specific conditions which can depend on region. This can be a probable cause of the disagreement between the observed and calculated trends in stratospheric  $NO_2$ .

For interpreting trends and solar activity effects in stratospheric NO<sub>2</sub>, three-dimensional models will be useful.

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Station	Latitude	Longitude	Institution	Observation
				period
1. Ny-Alesund	78.9 N	11.9 E	NILU	1991; 1993-2003
2. Scoresbysund	70.5 N	22.0 W	SA	1996-2007
3. Kiruna	67.8 N	20.4 E	NIWA	1991-2006
4. Sodankyla	67.5 N	26.6 E	SA	1995-2007
5. Zhigansk	66.8 N	123.4 E	SA	1992-2004; 2006-
				2007
6. Harestua	60.2 N	10.8 E	IASB	1994-2007
7. Zvenigorod	55.7 N	36.8 E	IAP	1990-2007
8. Jungfraujoch	46.6 N	8.0 E	IASB	1990-2007
9. Issyk Kul	42.6 N	77.0 E	KNU	1983-2005
10. Mauna Loa	19.5 N	155.6 W	NIWA	1996-2006
11. Bauru	22.4 S	49.0 W	SA	1996-2007
12. Lauder	45.0 S	169.7 E	NIWA	1981-2006
13. Kerguelen Isl.	49.4 S	70.3 E	SA	1996-2007
14. Macquarie Isl.	54.5 S	158.9 E	NIWA	1996-2004
15. Dumont d'Urville	66.7 S	140.0 E	SA	1988-2007
16. Rothera	67.6 S	68.1 W	BAS	1996-2005
17. Arrival Heights	77.8 S	166.7 E	NIWA	1991-2006

Table. Stations of NO<sub>2</sub> observations

NILU – Norwegian Institute for Air Research, Tromsø, Norway; DMI – Danish Meteorological Institute, Copenhagen, Denmark; SA – Service d'Aéronomie, CNRS, Paris, France; NIWA – National Institute of Water and Atmospheric Research, Lauder, New Zealand; IASB – Institut d'Aéronomie Spatiale de Belgique, Brusselex, Belgium;

IAP – A.M. Obukhov Institute of Atmospheric Physics, Moscow, Russia; RCAST – Research Centre for Advanced Science and Technology, Tokyo, Japan; KNU – Kyrgyz National University, Bishkek, Kyrgyzstan; BAS – British Antarctic Survey, Cambridge, UK; IU – Institut für Umweltphysik, Universität Heidelberg, Germany

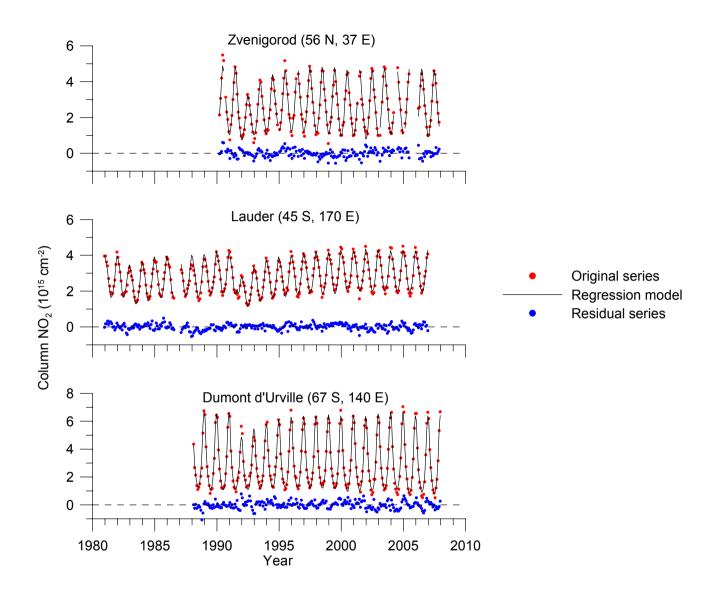
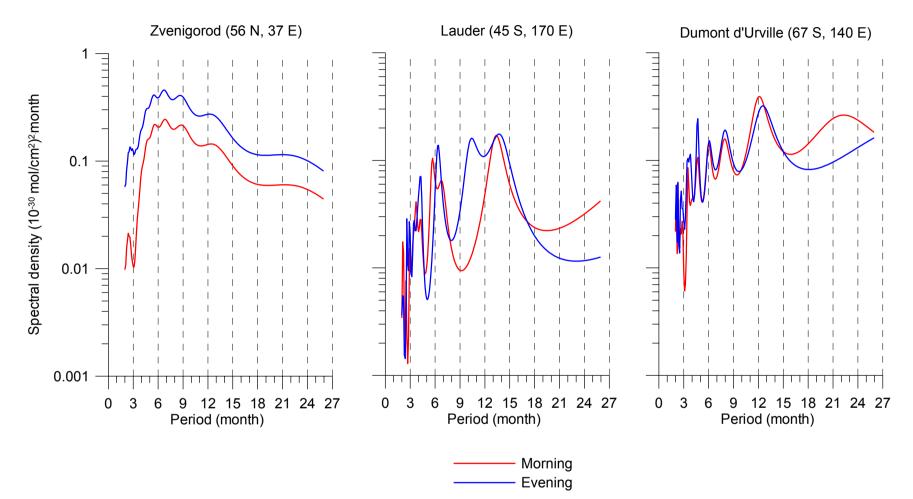


Fig. 1. Monthly means of stratospheric column NO<sub>2</sub>, their approximation with the regression model, and residual series according to morning measurements at Zvenigorod, Lauder, and Dumont d'Urville stations.



### Power spectra of residual series

Fig. 2. Power spectra of residual series.

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# Annual estimates of the linear trend

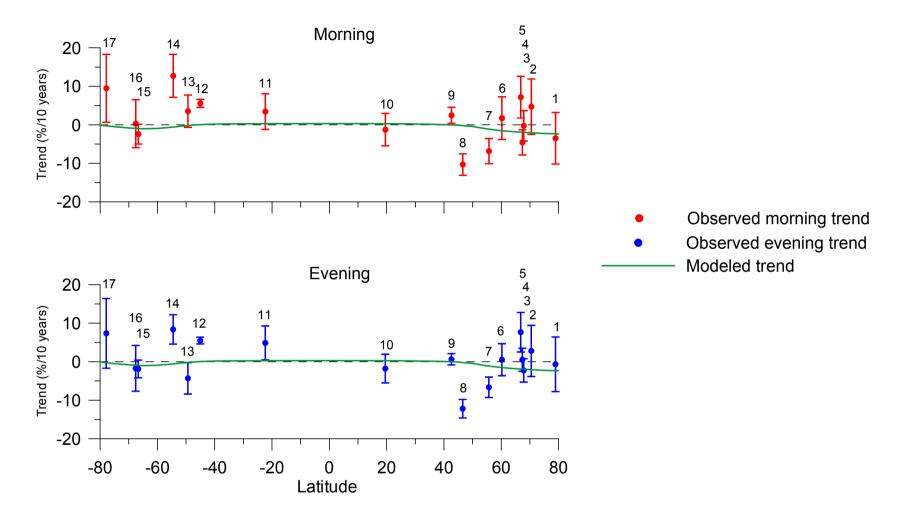


Fig. 3. Annual estimates of the stratospheric  $NO_2$  linear trends as function of latitude according to morning and evening measurements and SOCRATES model calculations. The vertical segments are 95% confidence intervals. The numbers are station numbers in Table.

# Seasonal trend estimates

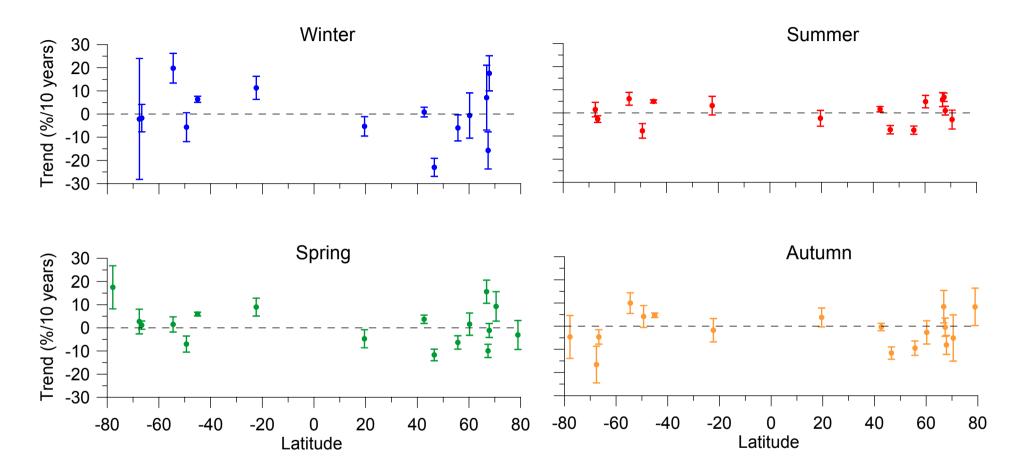


Fig. 4. Seasonal estimates of the stratospheric  $NO_2$  linear trends as function of latitude according to morning measurements. The vertical segments are 95% confidence intervals.

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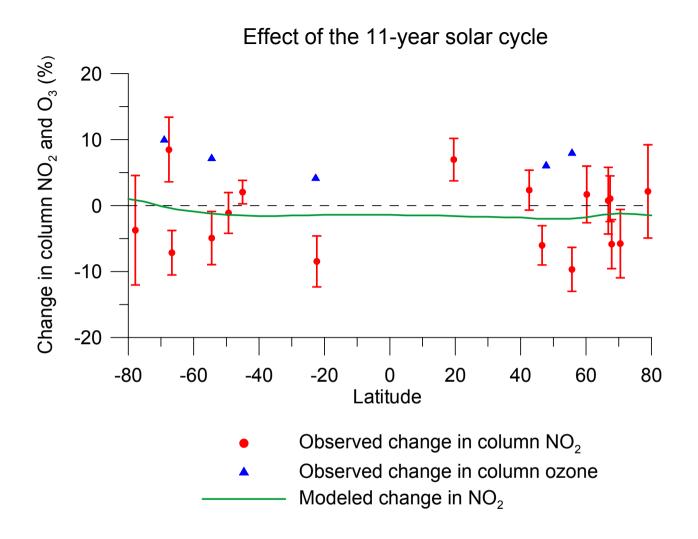


Fig. 5. Change in stratospheric column  $NO_2$  from the minimum to maximum phase of solar activity as function of latitude, according to morning measurements, and model (SOCRATES) calculations. The vertical segments are 95% confidence intervals.

## Pinatubo effect

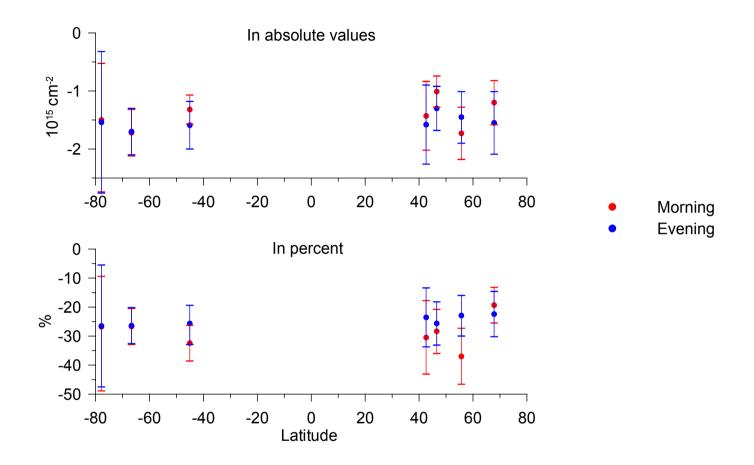


Fig. 6. Maximum values of the decrease in stratospheric column NO<sub>2</sub> after the Pinatubo eruption and their 95% confidence intervals according to morning and evening measurements as function of latitude, in absolute units and percentage with respect to long-term monthly means.

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